HARD1.071A PATENT

FIBER CEMENT COMPOSITE MATERIALS USING BLEACHED CELLULOSE FIBERS

Related Applications

[0001] This application claims the benefit of U.S. Provisional Application No. 60/439,040, filed January 9, 2003, the entirety of which is hereby incorporated by reference.

Background of the Invention

Field of the Invention

[0002] This invention relates generally to a fiber-reinforced composite material, and more particularly to a fiber cement building material utilizing a blend of bleached and unbleached cellulose fibers as reinforcing fibers, including formulations, methods of manufacture, and final products.

Description of the Related Art

[0003] Fiber-reinforced composite materials have been used to form various building products such as building sheets, panels, planks and roofing material. The reinforcement fibers used in these building products typically include natural and synthetic fibers such as metal fibers, glass fibers, asbestos fibers, and cellulose fibers such as those described in Australian Patent No. 515151 and U.S. Patent No. 6,030,447, which are hereby incorporated by reference in their entirety. Presently, cellulose fiber is one of the preferred fibers for most commercial building material applications because it is an effective, low cost, and recyclable natural product that is compatible with most conventional fiber cement manufacturing processes such as refining and autoclaving.

[0004] The performance characteristics of most cellulose fiber reinforced building materials are highly dependent on the quality and characteristic of the cellulose fibers used. In particular, the species and grade of cellulose fibers can have a significant impact on the flexibility and strength of the building material. For example, unbleached, premium grade cellulose fibers derived from *Pinus*. Radiata are known to provide the resulting product with higher strength because these fibers typically have longer length and favorable strength-related properties. Conversely, unbleached, standard grade cellulose fibers derived

from a variety of other commonly available wood species such as Douglas fir, hemlock, spruce, white fir, southern pine, and redwood are generally shorter and/or weaker, imparting lower strength and flexibility to the product.

[0005] However, there are several disadvantages associated with the use of premium grade cellulose fibers. First, the source of premium grade cellulose pulp has been largely limited to one particular species of wood. The cost of such fibers is also significantly higher than other grades of unbleached pulp. Moreover, limited availability of premium grade pulp can place severe constraints on fiber cement manufacturing operations and ultimately drive up the cost of the product. As such, product manufacturers typically incorporate both premium grade and standard grade unbleached pulps in fiber-cement composite materials in order to provide a final product with adequate strength and flexibility at minimum cost.

[0006] Even so, unbleached standard grade Kraft pulps typically used for fiber cement are considered specialty grades of cellulose pulp and are sold at a premium price over other types of cellulose pulp commonly used in paper, linerboard or other cellulose pulp based products.

[0007] Hence from the foregoing, it will be appreciated that there is a need for a readily available, less costly substitute for premium grade cellulose fibers for use in the manufacturing of fiber cement composite materials. To this end, there is a particular need for a cellulose fiber that will provide fiber cement composite materials with substantially equal if not improved strength, flexibility and other physical properties as compared to equivalent composite materials reinforced with premium grade cellulose fibers.

Summary of the Invention

[0008] The aforementioned needs are satisfied by the preferred embodiments of the present invention, which in certain embodiments disclose the novel concept of using a blend of bleached and unbleached standard grade cellulose fibers as a partial or complete substitute for premium grade cellulose fibers in the manufacture of fiber cement reinforced composite materials.

[0009] The terms "bleached cellulose fibers", "bleached fibers" and "bleached cellulose pulp" used herein refer to cellulose fibers that have been treated with a bleaching agent such as hydrogen peroxide, sodium hypochlorite, chlorine, and/or chlorine dioxide. The bleached cellulose fibers may encompass pulp derived from a variety of species of wood including but not limited to Douglas fir, hemlock, P. *Radiata* pine, white fir, spruce, southern yellow pine, kenaf, and redwood. The bleached cellulose fibers can be prepared by Kraft, Sulfite, or other pulping methods.

[0010] The terms "premium grade cellulose fibers", "premium grade fibers" and "premium grade cellulose pulp" used herein refer to cellulose fibers derived from P. Radiata pines having an average fiber length of more than about 1.5 mm and an average tensile strength of more than about 12.5 km by TAPPI method T231. The terms "standard grade cellulose fibers", "standard grade fibers" and "standard grade cellulose pulp" used herein refer to cellulose fibers derived from a variety of commonly available wood species such as Douglas fir, hemlock, spruce, white fir, southern pine, kenaf, and redwood, excluding P. Radiata pines. The term "standard grade cellulose fibers" can also be used to refer to "fiber cement grade" cellulose fibers known in the art.

[0011] In one aspect, the preferred embodiments of the present invention provide a composite material comprising a cementitious matrix and cellulose fibers incorporated into the cementitious matrix, wherein the cellulose fibers comprise a blend of bleached and unbleached cellulose fibers. In one embodiment, the bleached cellulose fibers comprise less than about 50%, preferably between about 5%-25%, of the total cellulose fibers incorporated into the matrix. In another embodiment, the bleached cellulose fibers have a Kappa number of less than or equal to about 10.

[0012] The bleached cellulose fibers are preferably standard grade fibers derived from species selected from the group consisting of Douglas fir, hemlock, spruce, southern pines, and redwood. In certain embodiments, the bleached cellulose fibers can also be premium grade fibers derived from R. Radiata pine. The unbleached cellulose fibers are preferably standard grade fibers comprising species selected from the group consisting of Douglas fir, hemlock, white fir, spruce, southern pine and redwood. In one embodiment, the bleached and unbleached cellulose fibers combined comprise about 0.5%-20% by weight of

the composite material. In another embodiment, the bleached and unbleached cellulose fibers comprise standard grade cellulose fibers having an average fiber length of between about 1 mm to 3.5 mm. Preferably, the modulus of rupture (MOR) and toughness energy of the composite material reinforced with the blend of bleached and unbleached fibers are substantially equal to or greater than that of an equivalent material reinforced with unbleached, premium grade cellulose fibers.

[0013] In another aspect, the preferred embodiments of the present invention provide a method of manufacturing a fiber-reinforced cement composite material. method comprises providing bleached and unbleached cellulose fibers; mixing the bleached and unbleached cellulose fibers with a cementitious binder to form a fiber cement mixture; forming the fiber cement mixture into a fiber cement article of a pre-selected shape and size; and curing the fiber cement article. In one embodiment, the step of providing the bleached cellulose fibers comprises treating standard grade cellulose fibers with a bleaching agent. Preferably, the bleached cellulose fibers have an average Kappa number of less than or equal to about 10. In another embodiment, providing the unbleached cellulose fibers comprises providing unbleached, standard grade cellulose fibers. Preferably, the bleached and unbleached cellulose fibers are mixed in pre-selected proportions to provide the composite material with pre-determined physical properties. In one embodiment, the pre-selected proportion is formulated to provide the composite material with a modulus of rupture (MOR) that is substantially equal to or greater than the MOR of an equivalent composite material reinforced with only premium grade cellulose fibers.

[0014] In yet another aspect, the preferred embodiments of the present invention provide a composite building material comprising a cementitious matrix and a blend of cellulose fibers. Preferably, the blend of cellulose fibers comprises bleached and unbleached cellulose fibers and is selected to provide the building material with pre-determined flexibility and strength. In one embodiment, the blend of cellulose fibers is selected to provide the building material with flexibility and tensile strength that are substantially equal or superior to that of an equivalent building material reinforced with only premium-grade cellulose fibers. In another embodiment, the blend of cellulose fibers is selected to provide the building material with flexibility and tensile strength that are substantially equal or

superior to the flexibility and strength of an equivalent building material reinforced with only bleached cellulose fibers. Preferably, the blend of cellulose fibers comprises less than about 50%, more preferably between about 5%-25%, bleached cellulose fibers. Moreover, the blend of cellulose fibers preferably does not include premium grade cellulose fibers.

[0015] In yet another aspect, the preferred embodiments of the present invention provide a composite material comprising a cementitious matrix, a first portion of cellulose fibers having a Kappa number of less than or equal to about 10 and a second portion of standard grade cellulose fibers having a Kappa number of greater than about 10. In one embodiment, the first portion of cellulose fibers comprises premium grade cellulose fibers. In another embodiment, the first portion of the cellulose fibers comprises less than about 50% by weight of the total amount of the two portions of the cellulose fibers combined. Advantageously, the bleached, standard grade cellulose fibers combine synergistically with the unbleached, standard grade fibers in the cementitious matrix, resulting in a composite material with substantially equal or improved strength and flexibility when compared with equivalent composite materials formed with only premium grade cellulose fibers.

Brief Description of the Drawings

- [0016] FIGURE 1 illustrates the process flow of a preferred method of forming a fiber cement composite material reinforced with a blend of bleached and unbleached cellulose fibers;
- [0017] FIGURE 2 illustrates the relationship between the modulus of rupture (MOR) of a fiber-cement composite material and varying amounts of bleached fibers incorporated in the material;
- [0018] FIGURE 3 illustrates the relationship between strain of the fiber-cement composite material and varying amounts of bleached fibers incorporated in the material;
- [0019] FIGURE 4 illustrates the relationship between toughness of the fiber-cement composite material and varying amounts of bleached fibers incorporated in the material.

Detailed Description of the Preferred Embodiment

[0020] Certain preferred embodiments of the present invention describe the use of a blend of bleached and unbleached cellulose fibers as reinforcement fibers in cementitious composite materials. In some preferred embodiments, the bleached and unbleached fibers are both standard grade fibers that in combination can serve as a partial or complete substitute for the more expensive and less abundant premium grade cellulose fibers. These embodiments encompass not only the composite materials formed with the blend of bleached and unbleached fibers, but also the formulation and methods of manufacture of the composite materials.

[0021] Bleached cellulose fibers have been used almost exclusively in the paper-making industry to provide white papers and paperboards. It is also widely recognized that the bleaching process degrades the fiber structure and weakens its reinforcing capabilities, making bleached fibers undesirable for fiber cement composite material applications. Such belief has been well documented in various technical publications. For example, in U.S. Patent No. 4,985,119 entitled "CELLULOSE FIBER-REINFORCED STRUCTURE", it indicates that "such bleaching [of the fibers] is not preferred due to cost and fiber degradation." Accordingly, it is against conventional wisdom to use bleached fibers as a reinforcing fiber for fiber cement composite materials because bleached fibers are generally believed to produce a fiber cement product that is weaker and more brittle.

[0022] However, contrary to conventional wisdom, Applicant has found that bleached cellulose fibers when used in proper proportions with unbleached, standard grade cellulose fibers can result in a fiber cement composite material with substantially equal or even superior flexibility, strength, and other physical properties when compared to an equivalent composite material reinforced by the more costly and less abundant premium grade cellulose fibers. It has been surprisingly found that proper blends of the selected bleached fibers and unbleached standard grade pulp give the final product a good balance of strength and flexibility.

[0023] Without wishing to be bound by a specific theory, Applicant believes that the bleached cellulose fibers have reduced lignin content and increased number of reactive

sites on the fiber surface as compared with equivalent cellulose fibers not treated with a bleaching agent. Applicant believes that this allows improved bonding of the fibers with the cementitious matrix and helps overcome the shorter fiber length and fiber brittleness normally associated with standard grade cellulose pulps. When used in predetermined amounts that complement the properties of unbleached pulp, Applicant has surprisingly found that bleached cellulose pulps are indeed very effective reinforcement fibers for cement composites.

[0022] One embodiment of the present invention provides a fiber cement composite material that incorporates bleached cellulose fibers in combination with unbleached standard grade cellulose fibers as reinforcing fibers. In one embodiment, the individual fiber length is between about 1 mm to 3.5 mm. Preferably, the bleached and unbleached fibers are used as a partial or complete substitute for the premium grade cellulose fibers that are commonly used in most fiber cement composite materials. The bleached cellulose fibers are preferably fibers treated with a bleaching agent such as hydrogen peroxide, sodium hypochlorite, or the like to reduce the lignin content of the fibers to a Kappa number of less than or equal to about 10. In one embodiment, the bleached fibers can comprise standard grade cellulose pulp of species including but not limited to Douglas fir, hemlock, white fir, spruce, kenaf, southern pines and redwood. Moreover, it will be appreciated that the preferred embodiments of the present invention are not limited to the use of bleached cellulose fibers as a substitute for premium grade fibers, but also include the use of all chemically treated fibers with substantially reduced lignin content and increased number of reaction sites on the fiber surface. Without wishing to be bound by theory, it is believed that the exposed reaction sites will increase the number of fiber-to-fiber and fiber-tocement bonding in the matrix, which in turn imparts strength to the material comparable to that achieved by stronger fibers.

[0024] In one embodiment, the bleached cellulose fibers are incorporated in a fiber cement matrix in combination with unbleached, standard grade pulp. The bleached fibers preferably comprise less than about 50%, more preferably between about 5%-25%, of the total cellulose fibers incorporated into the matrix. The combination of bleached cellulose fibers and unbleached standard grade cellulose pulp can be used in a variety of composite

materials all having different proportions of cementitious binder, aggregate, and cellulose fibers.

[0025] Most of the embodiments described herein can be encompassed by the following formulation:

- about 10%-80% cementitious binder (which, in certain embodiments, is selected from the group consisting of high alumina cement, lime, high phosphate cement, ground granulated blast furnace slag cement, and mixtures thereof);
- about 20%-80% aggregate (which, in certain embodiments, is selected from group consisting of ground silica, amorphous silica, micro silica, geothermal silica, diatomaceous earth, coal combustion fly ash, blast furnace slag, granulated slag, steel slag, mineral oxides, mineral hydroxides, clays, magnasite or dolomite, metal oxides and hydroxide, polymeric beads, and mixtures thereof);
- about 0.5%-20% cellulose fibers as comprising a combination of bleached standard grade cellulose fibers and unbleached standard grade cellulose fibers, and/or natural inorganic fibers, and/or synthetic fibers, wherein less than about 50% of the total cellulose fibers is bleached fibers;
- about 0%-80% density modifiers (which, in certain embodiments, is selected from the
 group consisting of plastic materials, expanded polystyrene or other foamed polymer
 materials, glass and ceramic materials, calcium silicate hydrates, microspheres and
 volcano ashes including perlite, pumice, shirasu basalt, zeolites in expanded forms, and
 mixtures thereof); and
- about 0%-10% additives (which, in certain embodiments, is selected from the group consisting of viscosity modifiers, fire retardants, waterproofing agents, silica fume, geothermal silica, thickeners, pigments, colorants, plasticizers, dispersants, forming agents, flocculent, drainage aids, wet and dry strength aids, silicone materials, aluminum powder, clay, kaolin, alumina trihydrate, mica, metakaolin, calcium carbonate, wollastonite, polymeric resin emulsion, and mixtures thereof).
- [0025] Figure 1 provides a schematic illustration of a process flow 100 of manufacturing a fiber cement composite material of one embodiment of the present invention. As shown in Figure 1, the process 100 begins with Step 102, which comprises

providing bleached and unbleached standard grade cellulose fibers. In one embodiment, the bleached cellulose fibers are less than about 50% of the combined weight of the bleached and unbleached fibers. In another embodiment, Step 102 comprises treating standard grade cellulose fibers with a bleaching agent to form bleached fibers having a Kappa number of less than about 10. The process 100 continues with Step 104, which comprises mixing the bleached and unbleached cellulose fibers with a cementitious binder and other ingredients to form a fiber cement mixture. Subsequently, the fiber cement mixture is formed into a fiber cement article of a pre-selected shape and size in Step 106. The fiber cement article is then cured to form the fiber cement reinforced composite building material in Step 108.

[0026] The advantages of incorporating a blend of bleached and unbleached standard grade cellulose fibers in a fiber cement composite matrix are numerous. They include the following:

- resulting composite material achieves a desired balance of strength and flexibility that is normally achievable only through incorporation of premium grade cellulose fibers;
- resulting composite material utilizes bleached cellulose fibers that can be obtained from
 a variety of commonly available wood species including but not limited to Douglas fir,
 hemlock, spruce, and redwood;
- bleached fibers require less energy to refine, reducing the product cost of the resulting
 composite material;

Example 1

[0027] Table 1 compares the mechanical properties of the fiber-reinforced cement composite materials made with equivalent formulations in which Formulation A incorporates bleached cellulose fibers and Formulation B incorporates premium cellulose fibers. The materials were produced using a Hatschek machine and the results are based on many samples collected over 1 week of production. It can be seen that formulations containing about 13% bleached pulp resulted in products with similar mechanical properties as when compared with products based on an equivalent formulation containing about 13% premium fiber of R. *Radiata* pine.

Table 1: Property Comparison of Fiber	For	mulations
Averaged Physical Properties	C 13% bleached pulp	D 13% premium <i>Radiata</i> pine
MOR (MPa)	10.39	10.29
Dry Density (Kg/m³)	1.25	1.25
Strain (µm/m)	12444	11961
Toughness (KJ/m³)	10.39	10.99

[0028] The base formulation for A and B is: about 35% Portland cement, about 57% ground silica and about 8% cellulose pulp. About 13% of the Formulation A cellulose pulp is bleached pulp and about 13% of the Formulation B cellulose pulp is *Radiata* pine. The remainder of the pulp for both formulations is standard grade Douglas fir Kraft pulp. Mechanical properties such as the modulus of rupture (MOR), strain, and toughness, are tested by three-point bending under the wet condition in accordance with ASTM (American Standard Test Method) C1185-98a entitled "Standard Test Methods for Sampling and Testing Non-Asbestos Fiber-Cement Flat Sheet, Roofing and Siding Shingles, and Clapboards."

Example 2

[0029] Specimens of fiber cement composite materials were made in accordance with Formulations C and D as shown in Table 2 below. Formulation C incorporated a blend of bleached and unbleached cellulose fibers. The fiber blend contained about 20% bleached fibers and about 80% unbleached fibers. Formulation D was the control, which utilized all unbleached fibers. It will be appreciated that the fiber cement formulations were selected for comparison purposes only and that a variety of other formulations can be used without departing from the scope of the present invention.

[0030] The oven dry densities of the specimens made in accordance with Formulations C and D were each about 1.25 g/cm³. The bleached fibers were Kraft pulp from Douglas fir with brightness of about 88, a Kappa number of 0 to 1 by TAPPI method

T236 and an average fiber length of about 2.4 mm. The unbleached fibers were conventional standard grade cellulose fibers with an average fiber length of about 2.6 mm and a Kappa number of 26. Both the bleached and unbleached fibers were refined to about 450 CSF (Canadian Standard Freeness) measured by TAPPI method T227.

Table 2: Formulations for Table 3 Test Results

Formulation	Hydraulic	Aggregate	Fiber,	Fiber
Identification	Binder			
	Portland	Silica	Bleached Fiber	Unbleached
	Cement	12 H 3 MM (12 2 17 18 1	Dieached Fiber	Fiber
С	35%	57%	1.6%	6.4%
	35%	57%	0.0%	8.0%
	3370	3770	0.070	0.076

[0031] Table 3 below provides an illustrative comparison of various mechanical and physical properties of fiber cement specimens made in accordance with formulations that incorporate a blend of bleached and unbleached cellulose fibers (Formulation C) and those that use conventional, unbleached cellulose fibers (Formulation D). The modulus of rupture (MOR), strain, and toughness are tested by three-point bending under the wet condition in accordance with ASTM (American Standard Test Method) C1185-98a entitled "Standard Test Methods for Sampling and Testing Non-Asbestos Fiber-Cement Flat Sheet, Roofing and Siding Shingles, and Clapboards."

Table 3: Property Comparison of Fiber Reinforced Cement

Composite Materials With and Without Bleached Cellulose Fibers

Physical Properties	Formulations		
	C (with 20% bleached fiber)	D (no bleached fiber)	
MOR (MPa)	11.23	11.09	
Dry Density (Kg/m³)	1.25	1.25	
Strain (μm/m)	13491	14292	
Toughness (KJ/m³)	19.26	20.62	

[0032] As shown in Table 3, key mechanical properties such as modulus of rupture (MOR) and ultimate strain are generally the same or slightly higher for specimens made with a blend of the bleached and unbleached fibers in accordance with Formulations C when compared to specimens made in accordance with Formulation D, the control formulation without bleached fibers. An equivalent formulation is herein defined as one in which the weight of unbleached cellulose fibers are displaced by an equivalent weight of bleached cellulose fibers. It will be appreciated that these are exemplifying results. By varying the proportional composition of bleached fibers, it will be appreciated that the physical and mechanical properties, such as MOR and strain, etc., of the final products can be changed to meet specific application needs.

[0033] Figures 2 to 4 illustrate the relationship between certain mechanical properties (MOR, strain and toughness) of the fiber-cement products and the percentage of bleached fibers in a fiber blend. It can be seen that percentage of bleached fibers is important for the fiber-cement composite materials to have a balanced properties of MOR, strain and toughness. Excessive bleached fibers present in the blend can adversely affect certain properties. For example, MOR will increase but the strain and toughness will decrease as the percentage of bleached fibers increases. In certain embodiments, to ensure good MOR as well as good strain and toughness, maximum proportions of bleached fibers shall not exceed 40% of the total fibers as shown in Figures 2 to 4.

[0034] Although the foregoing description of the preferred embodiments of the present invention has shown, described and pointed out the fundamental novel features of the invention, it will be understood that various omissions, substitutions, and changes in the form of the detail of the apparatus as illustrated as well as the uses thereof, may be made by those skilled in the art, without departing from the spirit of the invention. Consequently, the scope of the invention should not be limited to the foregoing discussions, but should be defined by the claims presented in the subsequently filed utility patent application.